Improving Electronic-Structure Calculations to (Machine-)Learn Chemistry

Elena Kolodzeiski

Technical University of Munich, TUM School of Natural Sciences, Department of Chemistry, Garching, 85748, Germany

Atomistic simulations have matured considerably over the last years with a plethora of applications ranging from the rational design of novel catalysts and functionalized materials to the study of operando changes in surface chemistry. In all cases, the prediction quality critically depends on a balanced description of molecular interactions independent of the interaction type and the length scale. In this talk, I will present our contributions to the method development and several examples where we successfully employed advanced computational methods in close connection with experiments to arrive at a detailed understanding of the underlying processes.

First, I will discuss the potential and limitations of density functional theory (DFT) calculations for the property prediction of carbon-based compounds like NHCs adsorbed to metal surfaces.[1-3] To address these limitations in terms of accuracy and computational requirements, we developed an embedding framework that efficiently reduces the computational cost and allows the systematic improvement of computational surface studies.[4] We validated this ACE-of-SPADE approach by benchmarking binding energies of CO_2 reduction products on a Cu(111) surface.[5] Secondly, I will present an on-the-fly training algorithm, which enables fast and automated parameterization of force fields for large-scale molecular dynamics simulations.[6-7] This is achieved by adapting and validating the force field continuously to automatically generated, accurate quantum-chemical reference data. The combination of the methodological developments allows us to bridge the gap between different time- and length scales and enables accurate predictions in many areas of chemical science, such as the development of novel homogeneous and heterogeneous catalysts, the study of soft matter, and the formation and behavior of nanoparticles.

(1) J. Ren; M. Das; H. Osthues; M. Nyenhuis; B. Schulze Lammers; E. Kolodzeiski; H. Mönig; S. Amirjalayer; H. Fuchs; N. L. Doltsinis; F. Glorius. *J. Am. Chem. Soc.* **2024**, 146 (11), 7288-7294.
(2) A. Bakker; M. Freitag; E. Kolodzeiski; P. Bellotti; A. Timmer; J. Ren; B. Schulze Lammers; D. Moock; H. W. Roesky; H. Mönig; S. Amirjalayer; H. Fuchs; F. Glorius. *Angew. Chem. Int. Ed.* **2020**, 59 (32), 13643-13646.
(3) A. Bakker; A. Timmer; E. Kolodzeiski; M. Freitag; H. Y. Gao; H. Mönig; S. Amirjalayer; F. Glorius; H. Fuchs. *J. Am. Chem. Soc.* **2018**, 140 (38), 11889-11892.
(4) E. Kolodzeiski; C. J. Stein. *J. Chem. Theory Comput.* **2023**, 19 (19), 6643-6655.
(5) E. Kolodzeiski; C. J. Stein. *in preparation* **2024**.
(6) E. Kolodzeiski; S. Amirjalayer. *J. Chem. Theory Comput.* **2021**, 17 (11), 7010-7020.
(7) E. Kolodzeiski; S. Amirjalayer. *Sci. Adv.* **2022**, 8 (26), eabn4426.